PHASE TRANSITIONS FOR BELOUSOV-ZHABOTINSKY REACTIONS

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Abstract. The main objective of this article is to study the dynamic phase transitions associated with the spatial-temporal oscillations of the BZ reactions, given by Field, Körös and Noyes, also referred as the Oregonator. Two criteria are derived to determine 1) existence of either multiple equilibria or spatiotemporal oscillations, and 2) the types of transitions. These criteria gives a complete characterization of the dynamic transitions of the BZ systems from the homogeneous states. The analysis is carried out using a dynamic transition theory developed recently by the authors, which has been successfully applied to a number of problems in science.

1. Introduction

In 1950's, in his experiments, B. P. Belousov discovered a spatial-temporal oscillation phenomenon in the concentrations of intermediaries when citric acid was oxidized by acid bromate in the presence of a cerium ion catalyst [1]. It is also observed by [14] that organic acids and metal ions could be used as well in the reaction, leading to spatial-temporal oscillations. It has been considered nowadays that all of the chemical reactions giving rise to oscillations, and the actions of catalyst are termed as the Belousov-Zhabotinsky (BZ) reactions. BZ reactions are now one of a class of reactions that serve as a classical example of non-equilibrium thermodynamics, resulting in the establishment of a nonlinear chemical oscillator.

The main objective of this article is to study the dynamic phase transitions associated with the spatial-temporal oscillations of the BZ reactions, given by Field, Körös and Noyes [2]. This BZ reaction consists of the following five irreversible steps:

$$
\begin{aligned}
&\mathrm{A} + \mathrm{Y} \xrightarrow{k_1} \mathrm{X},\\
&\mathrm{X} + \mathrm{Y} \xrightarrow{k_2} \mathrm{P},\\
&\mathrm{B} + \mathrm{X} \xrightarrow{k_3} 2\,\mathrm{X} + \mathrm{Z},\\
&2\,\mathrm{X} \xrightarrow{k_4} \mathrm{Q},\\
&\mathrm{Z} \xrightarrow{k_5} \gamma Y,
\end{aligned} \tag{1.1}
$$

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where γ is a stoichiometric factor, P and Q are products which do not join the reaction again, and

$$X = \mathrm{HBrO_2}, \quad Y = \mathrm{Br}^-, \quad Z = \mathrm{Ce}^{4+}, \quad A = B = \mathrm{BrO}_3^-.$$

It is the temporal oscillation of the curium ion ratio Ce(IV)/Ce(III) which, with a suitable indicator, is displayed by a color change, when the reagent is stirred.

The technical method for the analysis is the dynamic transition theory developed recently by the authors [6, 7, 8]. The main philosophy of the dynamic transition theory is to search for the full set of transition states, giving a complete characterization on stability and transition. The set of transition states is represented by a local attractor. Following this philosophy, the dynamic transition theory is developed to identify the transition states and to classify them both dynamically and physically. One important ingredient of this theory is the introduction of a dynamic classification scheme of phase transitions. With this classification scheme, phase transitions are classified into three types: Type-I, Type-II and Type-III, which, in more mathematically intuitive terms, are called continuous, jump and mixed transitions respectively. The dynamic transition theory is recently developed by the authors to identify the transition states and to classify them both dynamically and physically; see above references for details. The theory is motivated by phase transition problems in nonlinear sciences. Namely, the mathematical theory is developed under close links to the physics, and in return the theory is applied to the physical problems, although more applications are yet to be explored. With this theory, many long standing phase transition problems are either solved or become more accessible, providing new insights to both theoretical and experimental studies for the underlying physical problems.

With this method in our disposal, we derive in this article a characterization of dynamic transitions of the BZ reaction. In particular, the analysis in this article shows that the BZ system always undergoes a dynamic transition either to multiple equilibria or to periodic solutions (oscillations), dictated by the sign of a nondimensional computable parameter $\delta_0 - \delta_1$; see (3.12) and (3.16).

For the case of transitions to periodic solutions (spatiotemporal oscillations), the Type of transitions (Type-I and Type-II) are determined again by another computable nondimensional parameter. In the multiple equilibrium case, for general domains, the transition is always mixed (Type-III), while for rectangular domain, the transition is either continuous (Type-I) or jump (Type-II) based again on the sign of another nondimensional parameter. To demonstrate the applications, the derived characterization of dynamic transitions of the BZ system is then applied to a special example.

It is worth mentioning that continuous (Type-I) transitions imply that the concentrations will stay close to the basic homogeneous state, and the jump (Type-II) transition leads to more drastic changes in the concentrations. The mixed (Type-III) transition lead to two regions of initial concentrations corresponding to jump and continuous transitions respectively. In addition, both Type-II and Type-III transitions are accompanied with metastable states, and fluctuations between these metastable states; see [11] for the related concepts for binary systems.

This article is organized as follows. Section 2 introduces the basic model, and Section 3 study the dynamic transitions of the BZ model, and Section 4 gives an application of the theory and main results.

2. Field-Körös-Noyes equations

The stirred case was considered by [3], who derived a system of ordinary differential equations for this reaction. Here we consider the general cases. Let X, Y, Z be variable, and A, B be constants. The equations governing (1.1) are given by

$$\begin{aligned} \frac{\partial u_1}{\partial t} &= \sigma_1 \Delta u_1 + k_1 a u_2 - k_2 u_1 u_2 + k_3 b u_1 - 2k_4 u_1^2, \\ \frac{\partial u_2}{\partial t} &= \sigma_2 \Delta u_2 - k_1 a u_2 - k_2 u_1 u_2 + \gamma k_5 u_3, \\ \frac{\partial u_3}{\partial t} &= \sigma_3 \Delta u_3 + k_3 b u_1 - k_5 u_3, \end{aligned} \tag{2.1}$$

where u_1, u_2, u_3, a, b represent the concentrations of X, Y, Z, A, and B, σ_i $(i = 1, 2, 3)$ are the diffusivities of u_i, and k_j $(1 \le j \le 5)$ are the reaction coefficients as in (1.1). The model (2.1) is also called the Oregonator.

The coefficients σ_i and k_j are functions of the temperature T. In fact, $k_j = k_j^0 e^{-E_i/RT}$, E_i is the activation energy, and R is the Boltzmann constant.

The dimensions of the relevant quantities are given by:

$$\begin{aligned} &k_j: \ M^{-1} t^{-1} \text{ for } 1 \le j \le 4, && k_5: \ t^{-1}, \\ &a, b, u_i: \ M \text{ for } 1 \le i \le 3, && \sigma_i: \ l^2 t^{-1} \text{ for } 1 \le i \le 3 \end{aligned}$$

where t is the time, M is the mole density, and l is the length. Then we introduce the following nondimensional variables:

$$\begin{aligned} & u_1 = \frac{k_1 a}{k_2} u_1', && u_2 = \frac{k_3 b}{k_2} u_2', \\ & u_3 = \frac{k_1 k_3}{k_2 k_5} a b u_3', && t = (k_1 k_3 a b)^{-1/2} t', \\ & x = L x', && \alpha = \left(\frac{k_3 b}{k_1 a} \right)^{1/2}, \\ & \beta = \frac{2 k_1 k_4 a}{k_2 k_3 b}, && \delta = k_5 (k_1 k_3 a b)^{1/2}, \\ & \mu_i = \frac{\sigma_i}{l^2 (k_1 k_2 a b)^{1/2}} && \text{for } i = 1, 2, 3. \end{aligned}$$

Omitting the primes, we obtain the following nondimensional form of (2.1):

$$\begin{aligned} \frac{\partial u_1}{\partial t} &= \mu_1 \Delta u_1 + \alpha (u_1 + u_2 - u_1 u_2 - \beta u_1^2), \\ \frac{\partial u_2}{\partial t} &= \mu_2 \Delta u_2 + \frac{1}{\alpha} (\gamma u_3 - u_2 - u_1 u_2), \\ \frac{\partial u_3}{\partial t} &= \mu_3 \Delta u_3 + \delta (u_1 - u_3), \end{aligned} \tag{2.2}$$

where the unknown functions are $u_i \ge 0$ $(1 \le i \le 3)$, and the parameters are positive constants:

$$\mu_1, \ \mu_2, \ \mu_3, \ \alpha, \ \beta, \ \gamma, \delta > 0.$$

Let $\Omega \subset \mathbb{R}^n$, representing the container, be a bounded domain.

If there is an exchange of materials on the boundary $\partial\Omega$ to maintain the level of concentrations of X, Y, Z, then the equations (2.2) are supplemented with the

Dirichlet boundary condition

$$u = (u_1, u_2, u_3) = 0 \quad \text{on } \partial\Omega. \tag{2.3}$$

If there is no exchange of materials on the boundary, the equations are supplemented with the Neumann boundary condition

$$\frac{\partial u}{\partial n} = 0 \quad \text{on } \partial\Omega. \tag{2.4}$$

It is known in [12] that the following region

$$D = \{(u_1, u_2, u_3) \in L^2(\Omega)^3 \mid 0 < u_i < a_i,\ 1 \le i \le 3\} \tag{2.5}$$

is invariant for (2.2)-(2.3), where a_i $(1 \le i \le 3)$ satisfy

$$a_1 > \max\{1, \beta^{-1}\}, \quad a_2 > \gamma a_3, \quad a_3 > a_1.$$

The invariant region for the Neumann boundary condition case is the same as that of the Dirichlet boundary condition. For convenience, here we give the following lemma, which was well known in [12, 13]. This lemma shows that the model has a global attractor.

Lemma 2.1. *The region D given by (2.5) is also invariant for the problem (2.2) with (2.4). In particular this problem possesses a global attractor in D.*

Proof. We only need to prove that D is invariant for (2.2) with (2.4). It suffices to show that

$$\frac{\partial u}{\partial t} \text{ points inward at } x \in \partial D. \tag{2.6}$$

In fact, by (2.2), we see that

$$\begin{aligned}
&\frac{\partial u_1}{\partial t} = \alpha u_2 > 0 && \text{as } u_1 = 0,\ u_2 > 0,\ u_3 > 0,\\
&\frac{\partial u_2}{\partial t} = \frac{\gamma}{\alpha} u_3 > 0 && \text{as } u_2 = 0,\ u_2 > 0,\ u_3 > 0,\\
&\frac{\partial u_3}{\partial t} = \delta u_1 > 0 && \text{as } u_3 = 0,\ u_1 > 0,\ u_2 > 0,\\
&\frac{\partial u_1}{\partial t} = \alpha[a_1(1-\beta a_1) + (1-a_1)u_2] < 0 && \text{as } u_1 = a_1,\ u_2 > 0,\\
&\frac{\partial u_2}{\partial t} = -\frac{1}{\alpha}[(a_2 - \gamma u_3) + a_2 u_1] < 0 && \text{as } u_2 = a_2,\ u_1 > 0,\ 0 < u_3 < a_3,\\
&\frac{\partial u_3}{\partial t} = -\delta(a_3 - u_1) < 0 && \text{as } u_3 = a_3,\ 0 < u_1 < a_1,
\end{aligned}$$

where $a_1 > \max\{1, \beta^{-1}\}, a_2 > \gamma a_3$, and $a_3 > a_1$. These properties above show that (2.6) is satisfied. The lemma is proved. □

Let

$$\begin{aligned}
&\mathbb{R}^m_+ = \{(x_1, \cdots, x_m) \in \mathbb{R}^m \mid x_i \ge 0,\ 1 \le i \le m\},\\
&\lambda = (\mu_1, \mu_2, \mu_3, \alpha, \gamma, \delta) \in \mathbb{R}^6_+.
\end{aligned}$$

The we define the following function spaces:

$$H = L^2(\Omega)^3,$$

$$H_1 = \begin{cases} H^2(\Omega)^3 \cap H_0^1(\Omega)^3 & \text{for boundary condition (2.3)}, \\ \left\{ u \in H^2(\Omega)^3 \mid \dfrac{\partial u}{\partial n} = 0 \text{ on } \partial\Omega \right\} & \text{for boundary condition (2.4)}. \end{cases}$$

Define the operators $L_\lambda = A_\lambda + B_\lambda$ and $G_\lambda : H_1 \to H$ by

$$\begin{aligned} A_\lambda u &= (\mu_1 \Delta u_1, \mu_2 \Delta u_2, \mu_3 \Delta u_3), \\ B_\lambda u &= \left(\alpha u_1 + \alpha u_2, -\frac{1}{\alpha} u_2 + \frac{\gamma}{\alpha} u_3, \delta u_1 - \delta u_3 \right), \\ G(u, \lambda) &= \left(-\alpha(u_1 u_2 + \beta u_1^2), -\frac{1}{\alpha} u_1 u_2, 0 \right). \end{aligned}$$

Thus, the Field-Noyes equations (2.2), with either (2.3) or (2.4), take the following operator form:

$$\frac{du}{dt} = L_\lambda u + G(u, \lambda), \tag{2.7}$$

$$u(0) = \varphi \tag{2.8}$$

where $\lambda = (\mu_1, \mu_2, \mu_3, \alpha, \gamma, \delta) \in \mathbb{R}^6_+$.

We note that the solutions $u = (u_1, u_2, u_3)$ of (2.2) represent concentrations of chemical materials. Hence only the nonnegative functions $u_i \geq 0$ $(1 \leq i \leq 3)$ are chemically realistic.

3. Phase Transitions for BZ Reactions

3.1. **The model and basic states.** We consider case where there is no exchange of materials on the boundary. In this case, the model is supplemented with the Neumann boundary condition (2.4), and the system (2.2) admits two physically realistic constant steady state solutions:

$$U_0 = (0, 0, 0), \qquad U_1 = (u_1^0, u_2^0, u_3^0), \tag{3.1}$$

where

$$\begin{aligned} u_1^0 = u_3^0 = \sigma, \qquad u_2^0 &= \frac{\gamma\sigma}{1+\sigma} = \frac{1}{2}(1 + \gamma - \beta\sigma), \\ \sigma = \frac{1}{2\beta} &\left[(1 - \gamma - \beta) + \sqrt{(1 - \gamma - \beta)^2 + 4\beta(1+\gamma)} \right]. \end{aligned}$$

It is easy to check that the steady state solution $U_0 = 0$ is always unstable, because the linearized equations of (2.2) have three real eigenvalues λ_1, λ_2 and λ_3 satisfying

$$\lambda_1 \lambda_2 \lambda_3 = \det \begin{pmatrix} \alpha & \alpha & 0 \\ 0 & -\frac{1}{\alpha} & \frac{\gamma}{\alpha} \\ \delta & 0 & -\delta \end{pmatrix} = \delta(\gamma + 1) > 0,$$

and hence there always at least one eigenvalue with a positive real part.

Therefore we only need to consider the transition of (2.2) with (2.4) at the steady state solution U_1 in (3.1). For this purpose, we take the translation

$$u_i = u_i' + u_i^0 \quad (1 \leq i \leq 3). \tag{3.2}$$

Omitting the primes, the problem (2.2) with (2.4) becomes

$$
\begin{aligned}
&\frac{\partial u_1}{\partial t} = \mu_1 \Delta u_1 + \alpha[(1-u_2^0 - 2\beta u_1^0)u_1 + (1-u_1^0)u_2 - u_1u_2 - \beta u_1^2],\\
&\frac{\partial u_2}{\partial t} = \mu_2 \Delta u_2 + \frac{1}{\alpha}\left[-u_2^0 u_1 - (1+u_1^0)u_2 + \gamma u_3 - u_1u_2\right],\\
&\frac{\partial u_3}{\partial t} = \mu_3 \Delta u_3 + \delta(u_1 - u_3),\\
&\frac{\partial u}{\partial n}|_{\partial\Omega} = 0.
\end{aligned}
\tag{3.3}
$$

Then it suffices to study the phase transition of (3.3) at $u = 0$.

3.2. **Linear theory and principle of exchange of stabilities (PES).** In view of (3.1), the linearized eigenvalue equations of (3.3) are given by

$$
\begin{aligned}
&\mu_1\Delta u_1 - \alpha\left[\frac{1}{2}(\gamma + 3\beta\sigma - 1)u_1 + (\sigma - 1)u_2\right] = \lambda u_1,\\
&\mu_2\Delta u_2 - \frac{1}{\alpha}\left[\frac{1}{2}(1+\gamma-\beta\sigma)u_1 + (\sigma+1)u_2 - \gamma u_3\right] = \lambda u_2,\\
&\mu_3\Delta u_3 + \delta(u_1 - u_3) = \lambda u_3,\\
&\frac{\partial u}{\partial n}|_{\partial\Omega} = 0.
\end{aligned}
\tag{3.4}
$$

Let ρ_k and e_k be the kth eigenvalue and eigenvector of the Laplace operator Δ with the Neumann boundary condition:

$$
\begin{aligned}
&\Delta e_k = -\rho_k e_k, \qquad (\rho_k \geq 0),\\
&\frac{\partial e_k}{\partial n}|_{\partial\Omega} = 0.
\end{aligned}
\tag{3.5}
$$

Let M_k be the matrix given by

$$
M_k = \begin{pmatrix}
-\mu_1\rho_k - \frac{\alpha}{2}(\gamma + 3\beta\sigma - 1) & -\alpha(\sigma - 1) & 0\\
-\frac{1}{2\alpha}(1+\gamma-\beta\sigma) & -\mu_2\rho_k - \frac{1}{\alpha}(\sigma+1) & \frac{\gamma}{\alpha}\\
\delta & 0 & -\mu_3\rho_k - \delta
\end{pmatrix}.
\tag{3.6}
$$

Thus, all eigenvalues $\lambda = \beta_{kj}$ of (3.4) satisfy

$$
M_k x_{kj} = \beta_{kj} x_{kj}, \qquad 1 \leq j \leq 3,\ k = 1, 2, \cdots,
$$

where $x_{kj} \in \mathbb{R}^3$ is the eigenvector of M_k corresponding to β_{kj}. Hence, the eigenvector u_{kj} of (3.4) corresponding to β_{kj} is

$$
u_{kj}(x) = x_{kj} e_k(x),
\tag{3.7}
$$

where $e_k(x)$ is as in (3.5). In particular, $\rho_1 = 0$ and e_1 is a constant, and

$$
M_1 = \begin{pmatrix}
-\frac{\alpha}{2}(\gamma + 3\beta\sigma - 1) & -\alpha(\sigma - 1) & 0\\
-\frac{1}{2\alpha}(1+\gamma-\beta\sigma) & -\frac{1}{\alpha}(\sigma+1) & \frac{\gamma}{\alpha}\\
\delta & 0 & -\delta
\end{pmatrix}.
\tag{3.8}
$$

The eigenvalues $\lambda = \beta_{1j}$ $(1 \leq j \leq 3)$ of M_1 satisfy

$$
\begin{aligned}
&\lambda^3 + A\lambda^2 + B\lambda + C = 0, \\
&A = \delta + \left[\frac{\alpha}{2}(3\beta\sigma + \gamma - 1) + \frac{1}{\alpha}(\sigma + 1)\right], \\
&B = 2\beta\sigma^2 + \gamma - (1-\beta)\sigma + \delta\left[\frac{\alpha}{2}(3\beta\sigma + \gamma - 1) + \frac{1}{\alpha}(\sigma + 1)\right], \\
&C = \delta\sigma(2\beta\sigma + \beta + \gamma - 1).
\end{aligned} \tag{3.9}
$$

It is known that all solutions of (3.9) have negative real parts if and only if

$$
A > 0, \quad C > 0, \quad AB - C > 0. \tag{3.10}
$$

Direct calculation shows that these two parameters A and C in (3.9) are positive:

$$
A > 0, \quad C > 0; \tag{3.11}
$$

see also [4]. Note that

$$
\beta_{11}\beta_{12}\beta_{13} = -C < 0.
$$

It implies that all real eigenvalues of (3.8) do not change their sign, and at least one of these real eigenvalues is negative.

In addition, we derive, from $AB - C = 0$, the critical number

$$
\delta_0 = \frac{c + b - a^2 + \sqrt{(c + b - a^2)^2 + 4a^2 b}}{2a}, \tag{3.12}
$$

where

$$
\begin{aligned}
&a = \frac{\alpha}{2}(3\beta\sigma + \gamma - 1) + \frac{1}{\alpha}(\sigma + 1), \\
&b = (1-\beta)\sigma - 2\beta\sigma^2 - \gamma, \\
&c = \sigma(2\beta\sigma + \beta + \gamma - 1).
\end{aligned}
$$

It is then clear that $\delta_0 > 0$ if and only if

$$
b = (1-\beta)\sigma - 2\beta\sigma^2 - \gamma > 0, \tag{3.13}
$$

and under the condition (3.13)

$$
AB - C \begin{cases} > 0 & \text{if } \delta > \delta_0, \\ = 0 & \text{if } \delta = \delta_0, \\ < 0 & \text{if } \delta < \delta_0. \end{cases} \tag{3.14}
$$

Now we need to check the other eigenvalues β_{kj} with $k \geq 2$. By (3.6), $\lambda_k = \beta_{kj}$ $(k \geq 2)$ satisfy

$$
\lambda_k^3 + A_k\lambda_k^2 + B_k\lambda_k + C_k = 0, \tag{3.15}
$$

where

$$\begin{aligned}
A_k =& A + (\mu_1 + \mu_2 + \mu_3)\rho_k, \\
B_k =& B + (\mu_1 + \mu_2)\rho_k + a\mu_3\rho_k + \left(\frac{\sigma+1}{\alpha}\mu_1 + \frac{\alpha}{2}(3\beta\sigma| + \gamma - 1)\mu_2\right)\rho_k \\
&+ (\mu_1\mu_2 + \mu_1\mu_3 + \mu_2\mu_3)\rho_k^2, \\
C_k =& C + \left[\left(\frac{\sigma+1}{\alpha}\mu_1 + \frac{\alpha}{2}(3\beta\sigma + \gamma - 1)\mu_2\right)\delta - b\mu_3\right]\rho_k \\
&+ \left[\mu_1\mu_2\delta + \left(\frac{1+\sigma}{\alpha}\mu_1 + \frac{\alpha}{2}(3\beta\sigma + \gamma - 1)\mu_2\right)\mu_3\right]\rho_k^2 + \mu_1\mu_2\mu_3\rho_k^3,
\end{aligned}$$

where a, b are as in (3.12), and A, B, C are as in (3.9).

By (3.1) it is easy to check that

$$3\beta\sigma + \gamma - 1 > 0.$$

With the condition (3.13) we introduce another critical number

$$\delta_1 = \max_{\rho_k \neq 0}\left[\frac{\mu_3\rho_k b - C}{\left(\frac{\sigma+1}{\alpha}\mu_1 + \frac{\alpha}{2}(3\beta\sigma + \gamma - 1)\mu_2\right)\rho_k + \mu_1\mu_2\rho_k^2} - \mu_3\rho_k\right], \tag{3.16}$$

where b is as in (3.12).

Then the following lemma provides characterizes the PES for (3.4).

Lemma 3.1. *Let δ_0 and δ_1 be the numbers given by (3.12) and (3.16), and b given by (3.12). When $b < 0$, all eigenvalues of (3.4) always have negative real parts, and when $b > 0$ the following assertions hold true:*

(1) *Let $\delta_0 < \delta_1$ and $k_0 \geq 2$ the integer that δ_1 in (3.16) reaches its maximum at ρ_{k_0}. Then $\beta_{k_0 l}$ is a real eigenvalue of (3.4), and*

$$\beta_{k_1}(\delta)\begin{cases} < 0 & \text{if } \delta > \delta_1, \\ = 0 & \text{if } \delta = \delta_1, \\ > 0 & \text{if } \delta < \delta_1, \end{cases} \qquad \text{for } \rho_k = \rho_{k_0}$$

$$Re\beta_{ij}(\delta_1) < 0, \qquad \forall (i,j) \neq (k,1) \text{ with } \rho_k = \rho_{k_0}.$$

(2) *Let $\delta_0 > \delta_1$. Then $\beta_{11}(\delta) = \bar{\beta}_{12}(\delta)$ are a pair of complex eigenvalues of (3.4) near $\delta = \delta_0$, and*

$$Re\beta_{11} = Re\beta_{12}\begin{cases} < 0 & \text{if } \delta > \delta_0, \\ = 0 & \text{if } \delta = \delta_0, \\ > 0 & \text{if } \delta < \delta_0, \end{cases}$$

$$Re\beta_{kj}(\delta_0) < 0, \qquad \forall (k,j) \neq (1,1), (1,2).$$

Proof. In (3.9) we see that $A = a + \delta, B = a\delta - b$. By the direct calculation, we can see that

$$A_k > 0, \qquad A_kB_k - C_k > 0, \qquad \forall k \geq 2. \tag{3.17}$$

As $b < 0$, by (3.11)-(3.13),(3.15) and (3.17), for all physically sound parameters $\mu_1, \mu_2, \mu_3, \alpha, \beta, \delta > 0$, the following relations hold true

$$A_i > 0, \qquad C_i > 0, \qquad A_iB_i - C_i > 0, \qquad \forall i \geq 1.$$

Hence, it follows that all eigenvalues $\beta_{kj}(k \geq 1, 1 \leq j \leq 3)$ of (3.8) have negative real parts.

As $b > 0$, and $\delta_0 < \delta_1$, we infer from (3.11) and (3.14) that

$$\mathrm{Re}\beta_{1j}(\delta_1) < 0, \quad \forall 1 \leq j \leq 3. \tag{3.18}$$

In addition, it is clear that

$$\begin{aligned} &C_{k_1} \begin{cases} > 0 & \text{if } \delta > \delta_1, \\ = 0 & \text{if } \delta = \delta_1, \\ < 0 & \text{if } \delta < \delta_1, \end{cases} \\ &C_k > 0 \qquad \text{at } \delta = \delta_1 \text{ for all } k \neq k_1. \end{aligned} \tag{3.19}$$

Thus Assertion (1) follows from (3.17)-(3.19).

As $\delta_0 > \delta_1$, by (3.19) we know that $C_k > 0$ at $\delta = \delta_0$ for all $k \geq 2$. Since the real eigenvalues of β_{1j} $(1 \leq j \leq 3)$ are negative, the condition (3.14) implies that there are a pair of complex eigenvalues $\beta_{11} = \bar{\beta}_{12}$ cross the imaginary axis at $\delta = \delta_0$. Then Assertions (2) follows. The lemma is proved. $\square$

3.3. **Dynamic Phase Transitions.** By Lemma 3.1, as $\delta_1 < \delta_0$, the problem (3.3) undergoes a dynamic transition to a periodic solution from $\delta = \delta_0$. To determine the types of transition, we introduce a parameter as follows:

$$\begin{aligned} b_1 = &\frac{\rho^2}{D^2E}\Big[\frac{3}{D_0}((2D - 6 - \alpha\gamma\rho^2 D_3)F_3 - (2\gamma D_3 D_4 + \alpha D_5 D - 6)F_1) \\ &- \frac{1}{D_0}\big((\alpha\gamma\rho^2 D_3 - 2D_6)F_1 + (2\gamma D_3 D_4) + \alpha D_5 D_6)F_3\big) \\ &+ \frac{1}{D_0}(2\gamma\rho D_3 + \alpha\rho D_6 + \alpha\gamma\rho D_3 D_5 - 2\rho^{-1} D_4 D_6)F_2 \\ &+ \frac{\alpha}{2D^2E\rho}(2\gamma D_3 D_8 + \alpha D_6 D_7)(2\rho^{-1} D_6 D_8 - \alpha\gamma\rho D_3 D_7) \\ &+ \frac{\alpha\rho^2}{2D^2E}(2\gamma D_3 + \alpha D_6)(2\gamma D_3 D_8 + \alpha D_6 D_7) \\ &- \frac{\alpha\rho^2}{2D^2E}(2D_6 - \alpha\gamma\rho^2 D_3)(2\rho^{-2} D_6 D_8 - \alpha\gamma D_3 D_7)\Big], \end{aligned} \tag{3.20}$$

where

$$\rho = \sqrt{B} = \sqrt{\frac{C}{A}}, \qquad E = \alpha^3(\sigma-1)^3(\delta_0^2+\rho^2)^2,$$

$$D^2 = \frac{\gamma^2\delta_0^2\rho^2(\sigma-1)^2 + \rho^2((\delta_0^2+\rho^2)^2+\gamma\delta_0^2(\sigma-1))^2}{N^2(\sigma-1)^2(\delta^2+\rho^2)^4}, \qquad D_0 = \frac{\gamma\delta_0}{\alpha a^2} + \frac{a+2\delta_0}{\alpha(\delta-1)},$$

$$D_1 = \frac{\beta}{\alpha} + \frac{(\alpha A-2)(\beta\sigma+2\beta+\gamma-1)}{2\alpha(\sigma-1)^2}, \qquad D_2 = \frac{2-\alpha A}{\alpha^2(\sigma-1)^2},$$

$$D_4 = A+\alpha\beta\sigma^2+\alpha-3\alpha\beta\sigma-\alpha\beta-\alpha\gamma, \qquad D_3 = \delta_0(\sigma-1),$$

$$D_5 = A+\alpha-\alpha\beta\sigma-2\alpha\beta-\alpha\gamma, \qquad D_6 = (\delta_0^2+\rho^2)^2+\gamma\delta_0^2(\sigma-1),$$

$$D_7 = 1-\beta\sigma-\beta-\gamma, \qquad D_8 = \beta\sigma^2+\beta+1-3\beta\sigma-\gamma,$$

$$F_1 = \frac{D_1}{A} - \frac{2\rho^2 D_1}{A(A^2+4\rho^2)} - \frac{\rho D_2}{A^2+4\rho^2},$$

$$F_2 = \frac{D_2}{A} - \frac{4\rho^2 D_2}{A(A^2+4\rho^2)} + \frac{2\rho D_1}{A^2+4\rho^2},$$

$$F_3 = \frac{2\rho D_1}{A(A^2+4\rho^2)} + \frac{\rho D_2}{A^2+4\rho^2}$$

Here A, B, C are as in (3.9), and a is as in (3.12).

Then we have the following dynamic transition theorem.

Theorem 3.1. *Let $\delta_1 < \delta_0$, and b_1 is the number given by (3.20). Then the problem (3.3) undergoes a transition to periodic solutions at $\delta = \delta_0$, and the following assertions hold true:*

(1) *When $b_1 < 0$, the transition is of Type-I, and the system bifurcates to a periodic solution on $\delta < \delta_0$ which is an attractor.*
(2) *When $b_1 > 0$, the transition is of type-II, and the system bifurcates on $\delta > \delta_0$ to a periodic solution, which is a repeller.*

Proof. By Lemma 3.1, at $\delta = \delta_0$ there is a pair of imaginary eigenvalues $\beta_{11} = \bar{\beta}_{12} = -i\rho$ of (3.4). Let $z = \xi + i\eta$ and $z^* = \xi^* + i\eta^*$ be the eigenvectors and conjugate eigenvectors of (3.4) corresponding to $-i\rho$, i.e. z and z^* satisfy that

$$\begin{aligned} &(M_1 + i\rho)z = 0, \\ &(M_1^* - i\rho)z^* = 0, \end{aligned} \tag{3.21}$$

where M_1 is the matrix as in (3.8), and M_1^* the transpose of M. Because $\pm i\rho$ are solutions of (3.9), and $AB = C$ at $\delta = \delta_0$, we deduce that

$$\rho^2 = B = C/A. \tag{3.22}$$

For $z = (z_1, z_2, z_3)$, from the first equation of (3.21) we get

$$\begin{aligned} &\delta_0 z_1 = (\delta_0 - i\rho)z_3, \\ &\alpha(\sigma-1)z_2 = \left(-\tfrac{\alpha}{2}(\gamma+3\beta\sigma-1)+i\rho\right)z_1. \end{aligned} \tag{3.23}$$

Thus, we derive from (3.23) the eigenvectors $z = \xi + i\eta$ as follows:

$$\begin{aligned} &\xi = (\xi_1,\xi_2,\xi_3) = \left(1, -\tfrac{\gamma+3\beta\sigma-1}{2(\sigma-1)}, \tfrac{\delta_0^2}{\delta_0^2+\rho^2}\right). \\ &\eta = (\eta_1,\eta_2,\eta_3) = \left(0, \tfrac{\rho}{\alpha(\sigma-1)}, \tfrac{\rho\delta_0}{\delta_0^2+\rho^2}\right) \end{aligned} \tag{3.24}$$

In the same fashion, we derive from the second equation of (3.21) we derive the conjugate eigenvectors $z^* = \xi^* + i\eta^*$ as

$$\begin{aligned} \xi^* &= (\xi_1^*, \xi_2^*, \xi_3^*) = \left(-\tfrac{\sigma+1}{\alpha^2(\sigma-1)}, 1, \tfrac{\gamma\delta_0}{\alpha(\delta_0^2+\rho^2)}\right), \\ \eta^* &= (\eta_1^*, \eta_2^*, \eta_3^*) = \left(-\tfrac{\rho}{\alpha(\sigma-1)}, 0, -\tfrac{\gamma\rho}{\alpha(\delta_0^2+\rho^2)}\right). \end{aligned} \tag{3.25}$$

It is easy to show that

$$\begin{aligned} (\xi, \xi^*) &= (\eta, \eta^*) = -\tfrac{\gamma\rho^2 D_3}{H_1}, \\ (\xi, \eta^*) &= -(\eta, \xi^*) = -\tfrac{\rho D_6}{H_1}, \end{aligned} \tag{3.26}$$

where $H_1 = \alpha(\sigma-1)(\delta_0^2+\rho^2)^2, D_3$ and D_6 are as in (3.20). It is known that functions $\Psi_1^* + i\Psi_2^*$ given by

$$\begin{aligned} \Psi_1^* &= \tfrac{1}{(\xi,\xi^*)}[(\xi,\xi^*)\xi^* + (\xi,\eta^*)\eta^*], \\ \Psi_2^* &= \tfrac{1}{(\eta,\eta^*)}[(\eta,\xi^*)\xi^* + (\eta,\eta^*)\eta^*], \end{aligned} \tag{3.27}$$

also satisfy the second equation of (3.21) with

$$\begin{aligned} (\xi, \Psi_1^*) &= (\eta, \Psi_2^*) \neq 0, \\ (\xi, \Psi_2^*) &= (\eta, \Psi_1^*) = 0. \end{aligned} \tag{3.28}$$

Let $u \in H$ be a solution of (3.3) expressed as

$$u = x\xi + y\eta + \Phi(x,y), \qquad x, y \in \mathbb{R}^1.$$

where $\Phi(x,y)$ is the center manifold function of (3.3) at $\delta = \delta_0$. By (3.28), the reduced equations of (3.3) on the center manifold are given by

$$\begin{aligned} \frac{dx}{dt} &= -\rho y + \frac{1}{(\xi, \Psi_1^*)}(G(x\xi + y\eta + \Phi), \Psi_1^*), \\ \frac{dy}{dt} &= \rho x + \frac{1}{(\eta, \Psi_2^*)}(G(x\xi + y\eta + \Phi), \Psi_2^*), \end{aligned} \tag{3.29}$$

where $G(u) = G(u,u)$ is the bilinear operator defined by

$$G(u,v) = (-\alpha u_1 v_2 - \alpha\beta u_1 v_1, -\frac{1}{\alpha}u_1 v_2, 0) \tag{3.30}$$

for $u = (u_1, u_2, u_3), v = (v_1, v_2, v_3) \in H_1$.

We are now in position to solve the center manifold function $\Phi(x,y)$. To this end, we need to determine the third eigenvalue β_{13} and eigenvector ζ of M_1 at $\delta = \delta_0$. We know that

$$\beta_{13} \cdot (i\rho)(-i\rho) = -\rho^2\beta_{13} = \det M_1 = C.$$

By (3.22) we obtain

$$\beta_{13} = -A = -(\delta_0 + a),$$

and a is the number as in (3.12). Then, from the equation

$$(M_1 - \beta_{13})\zeta = 0,$$

we derive the eigenvector

$$\zeta = (\zeta_1, \zeta_2, \zeta_3) = \left(1, \frac{A - \frac{\alpha}{2}(\gamma + 3\beta\sigma + 1)}{\alpha(\sigma-1)}, -\frac{\delta_0}{a}\right). \tag{3.31}$$

In the same token, from

$$(M_1^* - \beta_{13})\zeta^* = 0,$$

we obtain the conjugate eigenvector as follows:

$$\zeta^* = (\zeta_1^*, \zeta_2^*, \zeta_3^*) = \left(\frac{A - \frac{1}{\alpha}(\sigma+1)}{\alpha(\sigma-1)}, 1, -\frac{\gamma}{\alpha a} \right). \tag{3.32}$$

On the other hand, from (3.24) and (3.30) it follows that

$$\begin{aligned} &G_{11} = G(\xi,\xi) = (-\alpha(\xi_2+\beta), -\tfrac{\xi_2}{\alpha}, 0), \\ &G_{12} = G(\xi,\eta) = (-\alpha\eta_2, -\tfrac{\eta_2}{\alpha}, 0), \\ &G_{22} = G(\eta,\eta) = 0, \\ &G_{21} = G(\eta,\xi) = 0. \end{aligned} \tag{3.33}$$

Direct calculation shows that

$$(\zeta,\zeta^*) = D_0, \qquad (G_{11},\zeta^*) = D_1, \qquad (G_{12},\zeta^*) = D_2,$$

and D_0, D_1, D_2 are as in (3.20).

By the approximation formula (A.11) in [10], the center manifold function Φ satisfy

$$\Phi = \Phi_1 + \Phi_2 + \Phi_3 + o(2), \tag{3.34}$$

where

$$\begin{aligned} &\mathcal{L}\Phi_1 = -x^2 P_2 G_{11} - xy P_2 G_{12}, \\ &(\mathcal{L}^2 + 4\rho^2)\mathcal{L}\Phi_2 = 2\rho^2(x^2-y^2)P_2G_{11} + 4\rho^2 xy P_2 G_{12}, \\ &(\mathcal{L}^2 + 4\rho^2)\Phi_3 = \rho(y^2-x^2)P_2G_{12} + 2\rho xy P_2 G_{11}, \end{aligned}$$

$P_2 : H \to E_2$ is the canonical projection, E_2 =the orthogonal complement of E_1=span$\{\xi,\eta)$, and $\mathcal{L}$ is the linearized operator of (3.3). By (3.7), it is clear that

$$\begin{aligned} &P_2G_{11} = (G_{11},\zeta^*)\zeta = D_1\zeta, \\ &P_2G_{12} = (G_{12},\zeta^*)\zeta = D_2\zeta. \end{aligned} \tag{3.35}$$

Hence $\Phi_1, \Phi_2, \Phi_3 \in \text{span}\{\zeta\}$. It implies that

$$\mathcal{L}\Phi_j = M_1\Phi_j = -A\Phi_j. \tag{3.36}$$

We infer from (3.34)-(3.36) the center manifold function as follows:

$$\begin{aligned} \Phi \;=\; & \frac{\zeta}{D_0}\Bigg[\left(\frac{D_1}{A} - \frac{2\rho^2 D_1}{A(A^2+4\rho^2)} - \frac{\rho D_2}{A^2+4\rho^2}\right)x^2 \\ & + \left(\frac{D_2}{A} - \frac{4\rho^2 D_2}{A(A^2+4\rho^2)} + \frac{2\rho D_1}{A^2+4\rho^2}\right)xy \\ & + \left(\frac{2\rho^2 D_1}{A(A^2+4\rho^2)} + \frac{\rho D_2}{A^2+4\rho^2}\right)y^2\Bigg] + o(2) \\ =\; & \frac{1}{D_0}(F_1x^2 + F_2xy + F_3y^2)\zeta + o(2), \end{aligned} \tag{3.37}$$

where F_1, F_2, F_3 are as in (3.21).

Inserting (3.37) into (3.29), by (3.27), we have

$$
\begin{aligned}
\frac{dx}{dt} =& -\rho y + \frac{1}{D^2}\big[[(\xi,\xi^*)(G_{11},\xi^*) + (\xi,\eta^*)(G_{11},\eta^*)]x^2 \\
&+ [(\xi,\xi^*)(G_{12},\xi^*) + (\xi,\eta^*)(G_{12},\eta^*)]xy \\
&+ \frac{1}{D_0}(\xi,\xi^*)(G(\xi,\zeta)+G(\zeta,\xi),\xi^*)(F_1x^3+F_2x^2y+F_3xy^2) \\
&+ \frac{1}{D_0}(\xi,\xi^*)(G(\zeta,\eta),\xi^*)(F_1x^2y+F_2xy^2+F_3y^3) \\
&+ \frac{1}{D_0}(\xi,\eta^*)(G(\xi,\zeta)+G(\zeta,\xi),\eta^*)(F_1x^3+F_2x^2y+F_3xy^2) \\
&+ \frac{1}{D_0}(\xi,\eta^*)(G(\zeta,\eta),\eta^*)(F_1x^2y+F_2xy^2+F_3y^3)\big] + o(3),
\end{aligned} \tag{3.38}
$$

$$
\begin{aligned}
\frac{dy}{dt} =& \rho x + \frac{1}{D^2}\big[[(\eta,\xi^*)(G_{11},\xi^*) + (\eta,\eta^*)(G_{11},\eta^*)]x^2 \\
&+ [(\eta,\xi^*)(G_{12},\xi^*) + (\eta,\eta^*)(G_{12},\eta^*)]xy \\
&+ \frac{1}{D_0}(\eta,\xi^*)(G(\xi,\zeta)+G(\zeta,\xi),\xi^*)(F_1x^3+F_2x^2y+F_3xy^2) \\
&+ \frac{1}{D_0}(\eta,\xi^*)(G(\zeta,\eta),\xi^*)(F_1x^2y+F_2xy^2+F_3y^3) \\
&+ \frac{1}{D_0}(\eta,\eta^*)(G(\xi,\zeta)+G(\zeta,\xi),\eta^*)(F_1x^3+F_2x^2y+F_3xy^2) \\
&+ \frac{1}{D_0}(\eta,\eta^*)(G(\zeta,\eta),\eta^*)(F_1x^2y+F_2xy^2+F_3y^3)\big] + o(3),
\end{aligned} \tag{3.39}
$$

where $D^2 = (\xi,\xi^*)^2 + (\xi,\eta^*)^2$.

Based on (3.24), (3.25), (3.30)-(3.33), we find

$$
\begin{aligned}
&(G_{11},\xi^*) = \frac{\alpha D_8}{H_2}, && (G_{12},\xi^*) = \frac{2\rho}{H_2}, \\
&(G_{11},\eta^*) = \frac{\alpha^2\rho D_7}{2H_2}, && (G_{12},\eta^*) = \frac{\alpha\rho^2}{H_2}, \\
&(G(\zeta,\eta),\xi^*) = \frac{2\rho}{H_2}, && (G(\zeta,\eta),\eta^*) = \frac{\alpha\rho^2}{H_2}, \\
&(G(\xi,\zeta)+G(\zeta,\xi),\xi^*) = \frac{2D_4}{H_2}, && (G(\xi,\zeta)+G(\zeta,\xi),\eta^*) = \frac{\alpha\rho D_5}{H_2}, \\
&H_2 = \alpha^2(\sigma-1)^2.
\end{aligned}
$$

In view of (3.26), equations (3.38) and (3.39) become

$$
\begin{aligned}
\frac{dx}{dt} =& -\rho y + \left[a_{20}x^2 + a_{11}xy + a_{30}x^3 + a_{21}x^2y + a_{12}xy^2 + a_{03}y^3\right] + o(3), \\
\frac{dy}{dt} =& \rho x + [b_{20}x^2 + b_{11}xy + b_{30}x^3 + b_{21}x^2y + b_{12}xy^2 + b_{03}y^3] + o(3),
\end{aligned}
$$

where

$$
\begin{aligned}
a_{20} &= -\frac{\rho^2}{2H_1H_2D^2}(2\alpha\gamma D_3D_8 + \alpha^2 D_6D_7),\\
a_{11} &= -\frac{\rho^3}{H_1H_2D^2}(2\gamma D_3 + \alpha D_6),\\
b_{20} &= \frac{\rho}{2H_1H_2D^2}(2\alpha D_6D_8 - \gamma\alpha^2\rho^2 D_3D_7),\\
b_{11} &= \frac{\rho^2}{2H_1H_2D^2}(2D_6 - \alpha\gamma\rho^2 D_3),\\
a_{30} &= \frac{\rho^2 F_1}{D_0H_1H_2D^2}(2\gamma D_3D_4 + \alpha D_5D_6),\\
a_{12} &= -\frac{\rho^2}{D_0H_1H_2D^2}[(2\gamma D_3D_4 + \alpha D_5D_6)F_3 + (2\gamma D_3 + \alpha D_6)\rho F_2]\\
b_{03} &= \frac{\rho^2 F_3}{D_0H_1H_2D^2}(2D_6 - \alpha\gamma\rho^2 D_3),\\
b_{21} &= \frac{\rho^2}{D_0H_1H_2D^2}[(2\rho^{-1}D_4D_6 - \alpha\gamma\rho D_3D_5)F_2 + (2D_6 - \alpha\gamma\rho^2 D_3)F_1]
\end{aligned}
$$

Due to Theorem A.3 in [5], the number

$$b_1 = 3(a_{30} + b_{03}) + (a_{12} + b_{21}) + \frac{2}{\rho}(a_{02}b_{02} - a_{20}b_{20}) + \frac{1}{\rho}(a_{11}a_{20} - b_{11}b_{20})$$

is the same as that given by (3.20). Thus the proof is complete. □

3.4. **Transition to steady state solutions.** Thanks to Lemma 3.1, for $\delta_1 > \delta_0$, the transition of (3.3) occurs at $\delta = \delta_1$, which is from real eigenvalues. Let δ_1 achieve it maximum at ρ_{k_0} ($k_0 \geq 2$). Assume that $\beta_{kol}(\delta)$ is simple near δ_1.

First, we consider the case where

$$\int_\Omega e_{k_0}^3 dx \neq 0. \tag{3.40}$$

In general, the condition (3.40) holds true for the case where $\Omega \neq (0, L) \times D$, and $D \subset \mathbb{R}^{n-1}(1 \leq n \leq 3)$ is a bounded open set.

Let

$$b_0 = [\alpha\beta(\sigma - 1)(\mu_2\rho_{k_0} + \frac{1}{\alpha}(\sigma + 1)) - (\alpha\mu_2\rho_{k_0} + 2\sigma)(\mu_1\rho_{k_0} + \frac{\alpha}{2}(\gamma + 3\beta\sigma - 1))].$$

Theorem 3.2. *Let the above number $b_0 \neq 0$, and $\delta_1 > \delta_0$. Under the condition (3.40), the transition of (3.3) at $\delta = \delta_1$ is mixed (Type-III), and the system bifurcates from $(u, \delta) = (0, \delta_1)$ to a unique steady state solution u^δ such that u^δ is a saddle on $\delta > \delta_1$, and an attractor on $\delta < \delta_1$. The solution u_δ can be expressed as*

$$u^\delta = C\beta_{k_0l}(\delta)\xi e_{k_0} + o(|\beta_{k_0|}|), \tag{3.41}$$

where $\xi = (\xi_1, \xi_2, \xi_3)$ *and constant* C *are given by*

$$\begin{aligned}
&\xi_1 = \alpha(\sigma-1)(\mu_3\rho_{k_0} + \delta_1),\\
&\xi_2 = -(\mu_1\rho_{k_0} + \frac{\alpha}{2}(\gamma + 3\beta\sigma - 1))(\mu_3\rho_{k_0} + \delta_1),\\
&\xi_3 = \alpha\delta_1(\sigma-1),\\
&C = \frac{[(\mu_3\rho_{k_0}+\delta_1)^2(\mu_1\rho_{k_0} + \mu_2\rho_{k_0} + \frac{1}{\alpha}(\sigma+1) + \frac{\alpha}{2}(\gamma+3\beta\sigma-1)) - (\sigma-1)\gamma\delta_1]\int_\Omega e_{k_0}dx}{b_0(\mu_3\rho_{k_0}+\delta_1)^3\int_\Omega e_{k_0}^3 dx}.
\end{aligned}$$

Proof. We apply Theorem A.2 in [9] to prove this theorem. Let ξ and $\xi^* \in \mathbb{R}^3$ be the eigenvectors of M_{k_0} and $M_{k_0}^*$ corresponding to $\beta_{k_0 l}(\delta_1) = 0$, i.e.

$$M_{k_0}\xi = 0, \qquad M_{k_0}^*\xi^* = 0,$$

where M_{k_0} is the matrix (3.6) with $k = k_0$. It is easy to see that ξ is as in (3.41), and

$$\begin{aligned}
&\xi^* = (\xi_1^*, \xi_2^*, \xi_3^*),\\
&\xi_1^* = -(\mu_2\rho_{k_0} + \frac{1}{\alpha}(\sigma+1))(\mu_3\rho_{k_0}+\delta_1),\\
&\xi_2^* = \alpha(\mu_3\rho_{k_0}+\delta_1)(\sigma-1),\\
&\xi_3^* = \gamma(\sigma-1).
\end{aligned} \tag{3.42}$$

For the operator G in (2.7), we can derive that

$$\frac{1}{(\xi e_{k_0}, \xi^* e_{k_0})}(G(y\xi e_{k_0}), \xi^* e_{k_0}) = -\frac{1}{C}y^2 + o(2),$$

where C is as in (3.41). Therefore, the theorem follows from Theorem A.2 in [9]. □

In the following, we assume that

$$\int_\Omega e_{k_0}^3 dx = 0.$$

Define

$$\begin{aligned}
b_1 = &\frac{1}{(\xi,\xi^*)}\big[-(\mu_1\rho_{k_0} + \frac{\alpha}{2}(\gamma+3\beta\sigma-1))(\alpha\mu_2\rho_{k_0}+2)\int_\Omega \phi_1 e_{k_0}^2 dx\\
&+ 2\alpha^2\beta(\sigma-1)(\mu_2\rho_{k_0} + \frac{\sigma+1}{\alpha})\int_\Omega \phi_1 e_{k_0}^2 dx\\
&+ \alpha(\sigma-1)(\alpha\mu_2\rho_{k_0}+2)\int_\Omega \phi_2 e_{k_0}^2 dx\big],
\end{aligned} \tag{3.43}$$

where

$$\begin{aligned}
(\xi,\xi^*) \quad = \quad & \alpha(1-\sigma)\big[(\mu_1\rho_{k_0} + \mu_2\rho_{k_0} + \frac{\sigma+1}{\alpha} + \frac{\alpha}{2}(\gamma+3\beta\sigma-1))\\
&\times(\mu_3\rho_{k_0}+\delta_1)^2 - \gamma\delta_1(\sigma-1)\big],
\end{aligned}$$

$\phi = (\phi_1, \phi_2, \phi_3)$ satisfies

$$L\phi = -G(\xi)e_{k_0}^2, \tag{3.44}$$

and the operators L and G are defined by

$$L\phi = \begin{cases} \mu_1\Delta\phi_1 - \alpha\left[\frac{1}{2}(\gamma+3\beta\sigma-1)\phi_1 + (\sigma-1)\phi_2\right], \\ \mu_2\Delta\phi_2 - \frac{1}{\alpha}\left[\frac{1}{2}(1+\gamma-\beta\sigma)\mu_1 + (\sigma+1)\phi_2\right], \\ \mu_3\Delta\phi_3 + \delta(\phi_1-\phi_3), \end{cases}$$

$$G(\xi) = \begin{cases} \alpha^2(\sigma-1)(\mu_3\rho_{k_0}+\delta_1)^2\left[\mu_1\rho_{k_0} + \frac{\alpha}{2}(\gamma+3\beta\sigma-1) - \alpha\beta(\sigma-1)\right], \\ (\sigma-1)(\mu_3\rho_{k_0}+\delta_1)^2(\mu_1\rho_{k_0} + \frac{\alpha}{2}(\gamma+3\beta\sigma-1)), \\ 0. \end{cases}$$

Then we have the following theorem.

Theorem 3.3. *Let $b_1 \neq 0$ be the number given by (3.43), and $\delta_0 < \delta_1$. Then the transition of (3.3) at $\delta = \delta_1$ is continuous (Type-I) as $b_1 < 0$, and is jump (Type-II) as $b_1 > 0$. Moreover, we have the following assertions:*

(1) *When $b_1 > 0$, this system bifurcates from $(u,\delta) = (0,\delta_1)$ to two steady state solutions u_+^δ and u_-^δ on $\delta > \delta_1$, which are saddles, and no bifurcation on $\delta < \delta_1$.*
(2) *When $b_1 < 0$, this system bifurcates to two steady state solutions u_+^δ and u_-^δ on $\delta < \delta_1$ which are attractors, and no bifurcation on $\delta > \delta_1$.*
(3) *The bifurcated solutions $u_\pm^\delta$ can be expressed as*

$$u_\pm^\delta = \pm\left[-\frac{\int_\Omega e_{k_0}^2 dx}{(\mu_2\rho_{k_0}+\delta_1)^2 b_1}\beta_{k_0 l}(\delta)\right]^{1/2} \xi e_{k_0} + o(|\beta_{k_0 l}|^{1/2}),$$

where $\xi \in \mathbb{R}^3$ is as in (3.41), $\beta_{k_0 l}(\delta)$ as in Lemma 3.1.

Proof. We use Theorem A.1 in [9] to verify this theorem. Let

$$u = y\xi e_{k_0} + \Phi(y), \qquad y \in \mathbb{R}^1,$$

and $\Phi(y)$ is the center manifold function. Let $\Phi = y^2\phi = o(2)$; then by the approximation formula of center manifolds (see (A.10) in [10]), ϕ satisfies

$$L\phi = -G(\xi e_{k_0}) = -G(\xi)e_{k_0}^2,$$

which is the equation (3.44).

We see that

$$\begin{aligned}(G(y\xi e_{k_0}+\Phi), \xi^* e_{k_0}) = &-\int_\Omega [\alpha(y\xi_1 e_{k_0}+\Phi_1)(y\xi_2 e_{k_0}+\Phi_2)\xi_1^* e_{k_0} \\ &+ \alpha\beta(y\xi_1 e_{k_0}+\Phi_1)^2\xi_1^* e_{k_0} + \frac{1}{\alpha}(y\xi_1 e_{k_0}+\Phi_1)(y\xi_2 e_{k_0}+\Phi_2)\xi_2^* e_{k_0}]dx \\ =&y^3[-(\alpha\xi_2\xi_1^* + 2\alpha\beta\xi_1\xi_1^* + \frac{1}{\alpha}\xi_2\xi_2^*)\int_\Omega \phi_1 e_{k_0}^2 dx \\ &- (\alpha\xi_1\xi_1^* + \frac{1}{\alpha}\xi_1\xi_2^*)\int_\Omega \phi_2 e_{k_0}^2 dx] + o(3)\end{aligned}$$

Hence, have

$$\frac{1}{(\xi e_{k_0}, \xi^* e_{k_0})}(G(y\xi e_{k_0}+\Phi), \xi^* e_{k_0}) = \frac{1}{\int_\Omega e_{k_0}^2 dx}(\mu_2\rho_{k_0}+\delta_1)^2 b_1 y^3 + o(y^3).$$

Thus the theorem follows from Theorem A.1 in [9]. □

3.5. **Stirred case.** Theorem 3.1 describes the spatial-temporal oscillation for the BZ reactions of (1.1) in a non-stirred condition. If the reagent is stirred, the equations (3.3) are reduced to the following system of ordinary differential equations:

$$\begin{aligned} \frac{du_1}{dt} &= -\alpha\left(\frac{1}{2}(\gamma+3\beta\sigma-1)u_1+(\sigma-1)u_2-u_1u_2-\beta u_1^2\right), \\ \frac{du_2}{dt} &= -\frac{1}{\alpha}\left(\frac{1}{2}(1+\gamma-\beta\sigma)u_1+(\sigma+1)u_2-\gamma u_3-u_1u_2\right), \\ \frac{du_3}{dt} &= \delta(u_1-u_3). \end{aligned} \tag{3.45}$$

In this case, only the transition to periodic solutions can take place, which is stated in the following theorem.

Theorem 3.4. *Let $\delta_0>0$ be the number given by (3.12), and b be as in (3.20). Then the system (3.45) undergoes a dynamic transition to periodic solutions at $\delta=\delta_0$. Furthermore, the system bifurcates to an unstable periodic orbit on $\delta>\delta_0$ as $b>0$, and to a stable periodic orbit on $\delta<\delta_0$ as $b<0$. In addition, the bifurcated periodic solution can be expressed in the following form*

$$u_\delta = [-b^{-1}Re\beta_{11}(\delta)]^{1/2}(\xi\cos\rho t+\eta\sin\rho t)+o(|Re\beta_{11}|^{1/2}), \tag{3.46}$$

where ξ,η are as in (3.32) and (3.34), and $\beta_{11}(\delta)$ is the first complex eigenvalue as described in Lemma 3.1.

Remark 3.1. Since the constant eigenvector space $E_1=\text{span}\{u_{11},u_{12},u_{13}\}=\mathbb{R}^3$ is invariant for (3.3) where u_{1j} are given by (3.7)), the bifurcated periodic solution u_λ in Theorem 3.1 has the same equations of (3.45) on the center manifold near $\delta=\delta_0$, given by

$$\begin{aligned} \frac{dx}{dt} &= \text{Re}\beta_{11}(\delta)x-\rho(\delta)y+\frac{1}{(\xi,\xi^*)}(G(x\xi+y\eta),\xi^*), \\ \frac{dy}{dt} &= \rho(\delta)x+\text{Re}\beta_{11}(\delta)x+\frac{1}{(\eta,\eta^*)}(G(x\xi+y\eta),\eta^*), \end{aligned} \tag{3.47}$$

where ξ^*,η^* are given by (3.35) and (3.36), $\rho(\delta)=\text{Im}\,\beta_{11}(\delta)$. The bifurcated periodic solution of (3.45) is written as

$$u_\delta = x(t)\xi+y(t)\eta+o(|x|,|y|).$$

In the polar coordinate system

$$x=r\cos\theta,\qquad y=r\sin\theta,$$

the solution $(x(t),y(t))$ of (3.47) can be expressed by

$$\begin{aligned} x(t) &= a(\delta)\cos\rho t+o(|a|), \\ y(t) &= a(\delta)\sim\rho t+o(|a|). \end{aligned}$$

The amplitude $a(\delta)$ satisfies

$$\begin{aligned} &\text{Re}\beta_{11}(\delta)+ba^2(\delta)+o(a^2)=0, \\ &a(\delta)\to 0 \text{ as } \delta\to\delta_0,\ a(\delta)>0. \end{aligned}$$

Thereby, we get

$$a(\delta)=[-b^{-1}\text{Re}\beta_{11}(\delta)]^{1/2}+o(|\text{Re}\beta_{11}|^{1/2}).$$

Thus, we get the expression (3.46).

Remark 3.2. By Lemma 2.1, for all physically-sound parameters, each of the two systems (3.3) and (3.45) has a global attractor in the invariant region D in (2.5). Hence, when $b > 0$, the bifurcated periodic solution u_δ is a repeller, and its stable manifold divides D into two disjoint open sets D_1 and D_2, i.e., $\bar{D} = \bar{D}_1 + \bar{D}_2, D_1 \cap D_2 = \emptyset$, such that the equilibrium $U_1 = (u_1^0, u_2^0, u_3^0)$ in (3.1) attracts D_1, and there is another attractor $\mathcal{A}_2 \subset D_2$ which attracts D_2.

4. An example

We begin with chemical parameters. In the chemical reaction (1.1), the constants given in [3] are as follows; see [4]:

$$
\begin{aligned}
&k_1 = 1.34M^{-1}S^{-1}, \qquad k_2 = 1.6\times 10^9 M^{-1}S^{-1}, \qquad k_3 = 8\times 10^3 M^{-1}S^{-1},\\
&k_4 = 4\times 10^7 M^{-1}S^{-1}, \qquad a = b = 6\times 10^{-2}M.
\end{aligned}
$$

Both γ and k_5 are order one parameters, and here we take:

$$\gamma = 1, \qquad k_5 = 1\cdot S^{-1},$$

and $a = b$ as a control parameter. The nondimensional parameters α, β, δ and μ_i $(1 \le i \le 3)$ are given by

$$
\begin{aligned}
&\alpha = \left(\frac{k_3 b}{k_1 a}\right)^{1/2} = 7.727\times 10,\\
&\beta = \frac{2k_1k_4a}{k_2k_3b} = 8.375\times 10^{-6},\\
&\delta = k_5(k_1k_3ab)^{1/2} = 1.035\times 10^2 aM^{-1},\\
&\mu_i = \frac{1}{(k_1k_2ab)^{\frac{1}{2}}}\frac{\sigma_i}{l^2} = 4.664\times 10^{-6}\frac{\sigma_i}{l^2 a}M^{-1}S^{-1},
\end{aligned}
\tag{4.1}
$$

for $i = 1, 2, 3$.

4.1. **Stirred case.** In view of (4.1), the numbers in (3.12) are as follows:

$$\sigma = 7\times 10^2, \qquad a = 9.74, \qquad b = 690.79, \qquad c = 8.21. \tag{4.2}$$

Hence

$$\delta_0 = 71.67. \tag{4.3}$$

Now we need to compute the parameter b_1 in (3.20). At the critical value $\delta_0 = 71.67$, the numbers in (3.9) are

$$A = a + \delta_0 = 81.41, \qquad B = a\delta_0 - b = 7.27, \qquad C = \delta_0 c = 588.41. \tag{4.4}$$

Then we have

$$
\begin{aligned}
&\rho = \sqrt{B} \cong 2.7, && E \cong 4\times 10^{21}, && D_0 \cong 1.3\times 10^{-2},\\
&F_1 \cong 8.3\times 10^{-9}, && F_2 \cong -2.6\times 10^{-7}, && F_3 \cong -8.4\times 10^{-10},\\
&D_3 \cong 5\times 10^4, && D_4 \cong 4\times 10^2, && D_5 \cong 80,\\
&D_6 \cong 3\times 10^7 && D_7 \cong -6\times 10^{-3}, && D_8 \cong 4.
\end{aligned}
\tag{4.5}
$$

Thus, by (4.1) and (4.2)-(4.5), the number b_1 in (3.20) is given by

$$\begin{aligned} b_1 \cong\ & \frac{\rho^2}{D^2 E D_0} \times \left[(\alpha\rho D_6 - 2\rho^{-1} D_4 D_6) F_2 - \alpha D_5 D_6 (3F_1 - F_3)\right] \\ & + \frac{\rho^2}{D^2 E}\left[\frac{\alpha^2 D_6^2 D_7 D_8}{E D^2 \rho^2} + \frac{\alpha^3 \rho^2 D_6^2 D_7}{2ED^2} - \frac{3.2\times 10^7 D_6 D_8}{\rho^2 E D^2}\right] \\ \cong\ & -\frac{\rho^2}{E D_0 D^2} \times 4 \times 10^3 - \frac{3.2 D_6 D_8}{E^2 D^4} \times 10^7 \\ & - \frac{6\alpha^2 D_6^2 D_8}{E^2 D^4} \times 10^{-3} - \frac{6\alpha^3 \rho^4 D_6^2}{2E^2 D^4} \times 10^{-3}. \end{aligned} \tag{4.6}$$

Hence, in the stirred situation, by Theorem 3.4 and (4.3)-(4.6), as $\delta < \delta_0 = 71.67$, the system (3.45) bifurcates from $(u, \delta) = (0, \delta_0)$ to a stable periodic solution, i.e. the reaction system (1.1) undergoes a temporal oscillation in the concentrations of $X = HBrO_2, Y = Br^-$, and $Z = Ce^{4+}$.

4.2. **Non-stirred case.** We consider the problem (3.3). In this case, there is another critical parameter δ_1 defined by (3.16). When $\delta_0 < \delta_1$, the system undergoes a transition to multiple equilibria at $\delta = \delta_1$, and when $\delta_0 > \delta_1$, the system has a transition to periodic solutions at $\delta = \delta_0$.

By (4.1) and (4.2), the number δ_1 is given by

$$\delta_1 = \max_{\rho_k}\left[\frac{690.8\mu_3}{9.1\mu_1 + 0.7\mu_2 + \mu_1\mu_2\rho_k} - \frac{588.4}{(9.1\mu_1 + 0.7\mu_2)\rho_k + \mu_1\mu_2\rho_k^2} - \mu_3\rho_k\right].$$

It is clear that if

$$\mu_3 \le \mu_1, \mu_2, \tag{4.7}$$

then in view of (4.3) we have

$$\delta_1 \le \frac{690.8\mu_3}{9.1\mu_1 + 0.7\mu_2} < \frac{690.8}{9.8} < \delta_0.$$

If (4.7) is not satisfied, let Ω_0 be a given domain, and Ω be the extension or contraction of Ω_0 from x_0 in the scale L $(0 < L < \infty)$ defined by

$$\Omega(L, x_0) = \{(x - x_0)L \mid x, x_0 \in \Omega_0, x_0 \text{ is fixed}\}. \tag{4.8}$$

In this case, the eigenvalues ρ_k $(k \ge 2)$ of (3.5) can be expressed by

$$\rho_k = \frac{C_k}{L^2} \quad \text{with } C_k > 0 \quad \text{and } C_k \to \infty \text{ as } k \to \infty.$$

It follows that there exists an $L_0 > 0$ such that $\delta_1 < \delta_0$ for any $L < L_0$. Thus, by Theorem 3.1 and (4.6), we have the following conclusion.

Physical Conclusion 4.1. *Under the condition of (4.7) or $\Omega = \Omega(L, x_0)$ with $L < L_0$, the system (3.3) bifurcates from $(u, \delta) = (0, \delta_0)$ to a stable periodic solution on $\delta < \delta_0$, which implies that the reaction system (1.1) with the Neumann boundary condition undergoes a temporal oscillation for $\delta < \delta_0$. However, when $\delta > \delta_0$, this system is in the trivial state U_1 in (3.1).*

Phase transition at $\delta = \delta_1$. By the above discussion, when $\mu_3 > \mu_i (i = 1, 2)$ and $L > L_0$ is large enough, the condition $\delta_0 < \delta_1$ may hold true. In the following, we assume that $L > L_0$ is sufficiently large, and

$$\mu_1 = \mu_2, \quad \mu_3 = z\mu_1 \quad (z > 1).$$

Then δ_1 becomes

$$\delta_1 \simeq \max_{x>0}\left[\frac{691z}{10+x} - \frac{589}{(10+x)x} - xz\right], \qquad (x = \mu_1\rho_k).$$

It follows that $3 < x < 4$ for $z > 1$. We take $x = \mu_1\rho_{k_0} = 3.5$, then

$$\delta_1 = 47.7z - 12.5$$

Thereby, we deduce that

$$\delta_1 > \delta_0 = 71.8 \quad \text{ as } \frac{\mu_3}{\mu_1} = z > 1.8.$$

Now, we investigate the transition of (3.3) at δ_1. For simplicity, we consider the case where

$$\Omega = (0, L) \subset \mathbb{R}^1.$$

In this case, the eigenvalues and eigenvectors of (3.5) are

$$\rho_k = (k-1)^2\pi^2/L^2; \quad e_k = \cos(k-1)\pi x/L.$$

It is clear that

$$\begin{aligned} & e_{k_0}^2 = (e_1 + e_j)/2 && \text{with } j = 2k_0 - 1, \\ & \int_\Omega e_{k_0}^3 dx = 0 && \text{by } k_0 \geq 2. \end{aligned}$$

We need to compute the number b_1 in (3.43). By (4.1), (4.2)-(4.3), and

$$\mu_3\rho_{k_0} = z\mu_1\rho_{k_0} = z\mu_2\rho_{k_0} = 3.5z \quad (z > 1.8),$$

the vectors $G(\xi)$ and ϕ in (3.44) are given by

$$\begin{aligned} & G(\xi) \approx (1.6\times 10^7(\mu_3\rho_{k_0} + \delta_1)^2, 3\times 10^3(\mu_3\rho_{k_0} + \delta_1)^2, 0), \\ & \phi = \varphi_0 e_1 + \varphi_j e_j \ (j = 2k_0 - 1), \\ & M_1\varphi_0 = -\frac{1}{2}G(\xi), \\ & M_j\varphi_j = -\frac{1}{2}G(\xi), \end{aligned}$$

where

$$\begin{aligned} M_1 &= \begin{pmatrix} -0.7 & -5.4\times 10^4 & 0 \\ -1.25\times 10^{-2} & -12.5 & 1.25\times 10^{-2} \\ \delta_1 & 0 & -\delta_1 \end{pmatrix}, \\ M_j &= \begin{pmatrix} -14.7 & -5.4\times 10^4 & 0 \\ -1.25\times 10^{-2} & -26.5 & 1.25\times 10^{-2} \\ \delta_1 & 0 & -(\mu_3\rho_j + \delta_1) \end{pmatrix}. \end{aligned}$$

The direct calculation shows that $\det M_1 = -8.75\delta_1, \det M_j \simeq -1.5\times 10^4 z + 4.9\times 10^3$, and

$$\begin{aligned} M_1^{-1} &= \frac{1}{\det M_1}\begin{pmatrix} 12.5\delta_1 & -5.4\times 10^4\delta_1 & * \\ 0 & 0.7\delta_1 & * \\ * & * & * \end{pmatrix}, \\ M_j^{-1} &= \frac{1}{\det M_j}\begin{pmatrix} 26.5(\mu_3\rho_j + \delta_1) & -5.4\times 10^4(\mu_3\rho_j + \delta_1) & * \\ -1.25\times 10^{-2}\mu_3\rho_j & 14.7(\mu_3\rho_j + \delta_1) & * \\ * & * & * \end{pmatrix}. \end{aligned}$$

Then we get

$$\begin{aligned}
&\varphi_0 = (\varphi_{01}, \varphi_{02}, \varphi_{03}) = -\frac{1}{2}M_1^{-1}G(\xi), && \varphi_{01} = 2.5 \times 10^6(\mu_3\rho_{k_0} + \delta_1)^2, \\
&\varphi_{02} = 1.1 \times 10^2(\mu_3\rho_{k_0} + \delta_1)^2, && \varphi_j = (\varphi_{j1}, \varphi_{j2}, \varphi_{j3}) = -\frac{1}{2}M_j^{-1}G(\xi) \\
&\varphi_{j1} = \frac{13.4 \times 10^3}{1.5z - 0.5}(\mu_3\rho_{k_0} + \delta_1)^2, && \varphi_{j2} = \frac{2z + 27}{1.5z - 0.5}(\mu_3\rho_{k_0} + \delta_1)^2.
\end{aligned}$$

Namely

$$\begin{aligned}
\phi_1 &= \varphi_{01}e_1 + \varphi_{j_1}e_j = (\mu_3\rho_{k_0} + \delta_1)^2\left[2.5 \times 10^6 e_1 + \frac{13.4 \times 10^3}{1.5z - 0.5}e_j\right], \\
\phi_2 &= \varphi_{02}e_1 + \varphi_{j2}e_j = (\mu_3\rho_{k_0} + \delta_1)^2\left[1.1 \times 10^2 e_1 + \frac{2z + 27}{1.5z - 0.5}e_j\right].
\end{aligned}$$

Putting ϕ_1 and ϕ_2 into (3.43), we derive that

$$\begin{aligned}
&b_1 = \frac{(\mu_3\rho_{k_0} + \delta_1)^2 L}{(\xi, \xi^*)}\left(5 \times 10^8 - \frac{5.7 \times 10^6}{1.5z - 0.5} - \frac{2z + 27}{1.5z - 0.5}\right), \\
&(\xi, \xi^*) = -699\alpha\left[16.7 \times (\delta_1 + 3.5z)^2 - 699\delta_1\right].
\end{aligned}$$

By $\delta_1 > \delta_0 = 71.8$ and $z > 1.8$ we see that $b_1 < 0$. Hence, by Theorem 3.3, the system (1.1) undergoes a dynamic transition at $\delta = \delta_1$ to the following steady state solutions

$$u_\pm = U_1 \pm \left(-\frac{L}{2(\mu_2\rho_{k_0} + \delta_1)^2 b_1}\beta_{k_0 l}(\delta)\right)^{1/2}\xi\cos\frac{(k_0 - 1)\pi}{L}x + o\left(\beta_{k_0 l}^{1/2}\right),$$

which are attractors.

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